

ELECTRICAL WIRE INSULATION

This invention relates to insulation for electrical wire or cable (hereinafter "wire") and is especially useful in multi-layer insulation of electrical wires

The following abbreviations will be used hereinafter:

PJ = Primary jacket; pro-rad = crosslinking promoter; TMPTM = trimethylolpropanetrimethacrylate; TAC = triallylcyanurate; TAIC = triallylisocyanurate; ASTM = American Society for Testing and Materials; PVDF = polyvinylidene fluoride; ETFE = ethylene/tetrafluoroethylene; ECTFE = ethylene/chlorotrifluoroethylene; HDPE = high density polyethylene; EEA = ethylene/ethyl acrylate; EA = ethyl acrylate; MFR = melt flow rate.

Wires with chemically-resistant dual-wall insulation comprising a polyolefin inner layer (core) and a fluoropolymer outer layer (primary jacket or PJ) have been commercially available for over 30 years, and are available from several different manufacturers. Dual wall wires which possess negligible bonding between these two insulation layers consequently suffer from a number of performance shortcomings, including a tendency to wrinkling, crack propagation, and peeling of the outer insulation layer. The tendency of polyolefin and fluoropolymer insulation layers not to bond appears to be at least partly due to the relatively high degree of chemical inertness of fluoropolymers. Indeed, in some cases, e.g. ethylene tetrafluoroethylene (ETFE), fluoropolymers are amongst the most chemically inert polymers known.

It has now been surprisingly discovered, according to the present invention, that the dissimilar insulation materials of a core based on a polyolefin and a PJ based on a fluoropolymer that is selected from a group of fluoropolymers which are hereinafter described as non-PVDF-based fluoropolymers (which term includes fully and (preferably) partially fluorinated fluoro-homopolymers and fluoro-copolymers and fluoro-terpolymers other than PVDF homopolymers or PVDF-based copolymers) can be bonded together to a significant level of adhesion on an electrical wire or cable, and that this bonding tends to reduce or eliminate outer insulation cracking on a wire and affords improvements in other performance

characteristics mentioned above. Significant bond strength is unexpectedly achieved by a combination of a selected formulation of a polyolefin-based layer, in contact with a non-PVDF-based fluoropolymer-based layer, and a cross-linking reaction, preferably effected by the application of radiation, especially ionising radiation.

The invention accordingly provides an electrical wire having insulation comprising:

(I) at least a first layer of a polyolefin-based material comprising at least 20%, preferably at least 40%, more preferably at least 60% or at least 80%, by weight (of the whole material composition) of a carbonyl-containing polymer (homopolymer or copolymer or terpolymer) having a non-aromatic backbone, of which polymer the or at least one constituent monomer is a carboxylic acid ester, preferably an acrylate or acetate, especially an alkyl acrylate (preferably methyl acrylate, ethyl acrylate, propyl acrylate or butyl acrylate), the said monomer itself constituting at least 5%, preferably at least 9%, more preferably at least 15% by weight of the said co-, or ter- polymer when used, and the remainder of the said co-, or ter-polymer preferably being derived from olefinic monomer, preferably ethylene;
in contact with;

(II) at least a second layer of a material containing at least 10%, more preferably at least 50%, or at least 90%, by weight based on the whole material composition, of a non-PVDF-based fluoropolymer or a mixture of such fluoropolymers, preferably a partially fluorinated non-PVDF-based fluoropolymer, preferably based on ETFE or on ECTFE;

wherein the said layers (I) and (II) whilst in contact with each other have been subjected to cross-linking reaction, preferably by radiation, more preferably ionising radiation, the cross-linking preferably being sufficient to increase the peel bond strength between the said layers to at least 3N (according to ASTM 1876-95), preferably increasing the bond strength by at least 50%, more preferably by at least 100%, especially by at least 500% or 1000%, compared to that between the layers prior to the crosslinking reaction.

Another aspect of the invention provides an electrical wire having insulation comprising:

(I) at least a first layer of a polyolefin-based formulation, of which at least 20%, preferably at least 40%, more preferably at least 60% or very preferably at least 80% of the weight of the polymeric portion of the said formulation consists of a carbonyl-containing polymer (homopolymer or copolymer or terpolymer), of which polymer the or at least one constituent

monomer is a carboxylic acid ester, preferably an acrylate or acetate, especially an alkyl acrylate (preferably methyl acrylate, ethyl acrylate, propyl acrylate or butyl acrylate), the said monomer itself constituting at least 5%, preferably at least 9%, more preferably at least 15% by weight of the said co-, or ter- polymer when used, and the remainder or the majority of the remainder of the said co-, or ter- polymer preferably being derived from olefinic monomer, preferably ethylene;

in contact with

(II) at least a second layer of a material containing at least 10%, more preferably at least 50%, or at least 90%, by weight based on the whole material composition, of a non-PVDF-based fluoropolymer or a mixture of such fluoropolymers, preferably a partially fluorinated non-PVDF-based fluoropolymer, preferably based on ETFE or on ECTFE or terpolymers based on ETFE or ECTFE with partially or fully fluorinated comonomers;

wherein the said layers (I) and (II) whilst in contact with each other have been subjected to cross-linking reaction, preferably by radiation, more preferably ionising radiation, the cross-linking preferably being sufficient to prevent delamination of the two layers during subsequent handling operations e.g. wrapping the wire round a mandrel of diameter equal to twice that of the insulated wire or to increase the peel bond strength between the said layers to at least 3N according to the ASTM B1876-95 method described below preferably increasing the bond strength by at least 50%, more preferably by at least 100%, especially by at least 500% or 1000%, compared to that between the layers prior to the cross-linking reaction.

Another aspect of the invention provides an electrical wire having insulation comprising:

(I) at least a first layer of a polyolefin-based formulation, of which at least 20%, preferably at least 40%, more preferably at least 60% or very preferably at least 80% of the weight of the polymeric portion of the said formulation consists of a carbonyl-containing polymer (homopolymer or copolymer or terpolymer), of which polymer the or at least one constituent monomer is a carboxylic acid ester, preferably an acrylate or acetate, especially an alkyl acrylate (preferably methyl acrylate, ethyl acrylate, propyl acrylate or butyl acrylate), the said monomer itself constituting at least 5%, preferably at least 9%, more preferably at least 15% by weight of the said co-, or ter- polymer when used, and the remainder or the majority of the remainder of the said co-, or ter- polymer preferably being derived from olefinic monomer, preferably ethylene;

in contact with

(II) at least a second layer of another material formulation, containing at least 10%, more preferably at least 50%, very preferably at least 90%, especially 100%, by weight of the second layer, of a non-PVDF-based ter-fluoropolymer, or a mixture of non-PVDF-based fluoropolymers containing one or more non-PVDF-based ter-fluoropolymers, preferably a partially fluorinated ter-fluoropolymer, more preferably terpolymers based on tetrafluoroethylene and hexafluoropropylene with fully or preferably partially or non-fluorinated comonomers such as ethylene or vinylidene fluoride, provided that the vinylidene fluoride comonomer does not constitute greater than 50wt% of the copolymer; wherein the said layers (I) and (II) whilst in contact with each other have been subjected to cross-linking reaction, preferably by radiation, more preferably ionising radiation, the cross-linking preferably being sufficient to prevent delamination of the two layers during subsequent handling operations e.g. wrapping the wire round a mandrel of diameter equal to twice that of the insulated wire or to increase the peel bond strength between the said layers to at least 3N according to the ASTM B1876-95 method described below preferably increasing the bond strength by at least 50%, more preferably by at least 100%, especially by at least 500% or 1000%, compared to that between the layers prior to the cross-linking reaction.

Preferably, the respective layers have been brought into contact with each other at a temperature above the melting or softening point of the polymeric material in at least one of the layers, thus tending to maximise the intimacy of their interfacial contact and so possibly encouraging the formation of adhesion-promoting interfacial cross-links in the subsequent cross-linking reaction.

The polyolefin-based layer (I) in addition to the polymeric portion of the formulation, for which the requirements are stipulated above, may contain whatever else is required in the way of additives such as anti-oxidants, pigments, fillers, flame retardants, etc, as known per se, to give the required mechanical, thermal, electrical etc. properties to the polymer.

The fluoropolymer-based layer (II) also may contain mixtures of two or more non-PVDF-based fluoropolymers and may contain other additives as known per se to give it required properties in addition to bonding.

Advantages of achieving a strong bond in accordance with this invention include:

- abrasion resistance of surface layer, and the insulation as a whole can increase if it (the surface layer) is bonded to a substrate material;
- improved resistance to peel, especially if one of the layers is damaged/perforated;
- improved resistance to blistering of the two layers, if heat is applied;
- improved resistance to delamination/creasing/wrinkling between the two layers, e.g. due to mechanical stress or chemical exposure e.g. to solvents.
- achievement of reduced wire bend wrinkling and improvement in the above characteristics, while maintaining adequate cut-through and notch propagation resistance, the latter being unexpected since strongly adherent layers would normally be expected fairly easily to transmit a cut or notch in the outer layer through to the inner layer.

The bond strength described in this application can be measured in terms of peel strength between bonded strips of the two materials in question. A standard method which can be used for such a test is ASTM 1876-95. By this definition, a moderate bond might have a peel force of >3N, a significant bond could be one for which the peel force exceeds 5N, and a strong bond one of peel force greater than 10N. A convenient method for gauging the bond strength between the said layers is to slit the insulation along the wire axis for 50mm, to the depth of the conductor, and to then wrap the wire round a mandrel, of diameter equal to twice the wire diameter, with the slit on the outside of the bend, so as to expose the conductor, and to then unwrap the wire from the mandrel until the wire is straight. If the inner and outer insulation layers have not spontaneously separated by this process there is at least a moderate bond between the layers.

Methods of fabricating the wire may include any process which causes intimate contact between the above-mentioned layers (i) and (ii). Examples include coating of one material onto a pre-formed layer of the other, dual or multi-walled extrusion to form insulation layers respectively containing one or other of the aforementioned two classes of material. The olefin-based material (i) is preferably the inner layer and the fluoropolymer-based layer (ii) preferably the outer layer on the wire. The layers made from the two different materials could be coextruded, tandem extruded, multipass extruded, or coated by other means. Known wire

insulation processes such as tube draw-down extrusion may be used, to form one or more of the layers, but pressure extrusion as known per se is preferred for optimum adhesion of the second and any subsequent insulation layers to be applied to a pre-formed underlying layer.

The insulation on the wire is exposed to a cross-linking reaction, which may involve chemical reagents such as peroxides, but preferably is effected by radiation, especially from a source of ionising radiation capable of causing the formation of free radicals and thus, cross-links, in the polymers, some of which should preferably be formed in the region of the interface between the two materials. Penetration of the radiation into the material at least as far as the interface is therefore desirable, although not necessarily essential if ion or radical mobility, for example, enables molecular reactions to continue at or near the interface after the radiation process. The radiation source could, for example, be a radio-isotope, or an X-ray source, or possibly a non-ionising radical-generating source, for example a UV source, but is preferably an electron beam, more preferably one providing a beam dose greater than 2 Mrads, preferably at least 5 Mrads, more preferably at least 10 Mrads, very preferably at least 15Mrads, into the material.

It has been found that enhancements to the interfacial bond strength may be obtained by using certain additives. Additives preferably include a cross-linking promoter ("pro-rad") in the polyolefin-based material and/or in the fluoropolymer-based material. Known cross-linking materials may be used, preferably methacrylate/acrylate based ones, and, very preferably, those of the type trimethylolpropanetrimethacrylate (TMPTM) in the polyolefin material and/or in the fluoropolymer-based material. Additionally, crosslinking promoters of the multifunctional allyl type, preferably triallyl cyanurate (TAC), more preferably triallyl isocyanurate (TAIC), in the polyolefin material and preferably in the fluoropolymer-based material, may be used.

Experimental results:

All results quoted in the tables below were obtained by testing pressed plaques of the two materials prepared by the usual polymer handling techniques, well known per se. The plaques were pressed together to adhere them temporarily face-to-face and the adhering plaques were then irradiated as indicated. Plaques were used to represent the layers I and II for these

demonstration experiments, due to the relative ease of measuring bond strength on plaques, rather than on wires. Conditions for these experiments were as follows:

Plaque dimensions: 150mm by 150mm by 0.85mm Pressing temperature: 220°C

Pressing time: 2 minute preheat, 1 minute at pressure

Pressing pressure: 20-40 Tons between 300mm by 300mm metal plates

Cooling conditions: 2 minutes at same pressure with same plates water cooled.

Example of Effect of Radiation Dose on Bond strength developed between appropriate polyolefin and fluoropolymer-based materials

Material 1	Material 2	Dose (Mrads)	Mean maximum peel force (N)
EEA copolymer of 15wt% EA content	ETFE of MFR*=33	0	0.5
EEA copolymer of 15wt% EA content	ETFE of MFR*=33	20	9
EMA copolymer of 28wt% MA content	ETFE of MFR*=33	0	0.8
EMA copolymer of 28wt% MA content	ETFE of MFR*=33	20	5
EEA copolymer of 15wt% EA content	ETFE of MFR*=5.5 + 5wt% TAC	0	0.5
EEA copolymer of 15wt% EA content	ETFE of MFR*=5.5 + 5wt% TAC	20	17.5
EEA copolymer of 15wt% EA content	ECTFE of MFR**=11	0	1.5
EEA copolymer of 15wt% EA content	ECTFE of MFR**=11	20	28
EEA copolymer of 15wt% EA content	Terpolymer of tetrafluoroethylene, hexafluoropropylene & vinylidene fluoride of MFR**=13	0	1.8
EEA copolymer of 15wt% EA content	Terpolymer of tetrafluoroethylene, hexafluoropropylene & vinylidene fluoride of MFR**=13	20	36
EEA copolymer of 15wt% EA content	Terpolymer of hexafluoropropylene, tetrafluoroethylene & ethylene of MFR**=6.7	0	2.4
EEA copolymer of 15wt% EA content	Terpolymer of hexafluoropropylene, tetrafluoroethylene & ethylene of MFR**=6.7	20	13

Example of Effect of the addition of Pro-rad in fluoropolymer-based material on bond strength with appropriate polyolefin-based material after electron beam crosslinking

Material 1	Material 2	Dose (Mrads)	Mean maximum peel force (N)
EEA copolymer of 15wt% EA content	ETFE of MFR*=5.5	20	6.5
EEA copolymer of 15wt% EA content	ETFE of MFR*=5.5 + 5wt% TAC	20	17.5

Effect of percentage Copolymer in a polyolefin polymer blend on bond strength with appropriate non-PVDF-based fluoropolymer-based material after electron beam crosslinking

Material 1	Material 2	Dose (Mrads)	Mean maximum peel force (N)
100% HDPE	ETFE of MFR*=33	20	0
100% EEA copolymer of 15wt% EA content	ETFE of MFR*=33	20	9

The Melt Flow Rate (MFR) used to describe the fluoropolymers in the tables above is quoted in units of grammes/10minutes and was measured according to ASTM D1238-95 under the following conditions; *297°C/5Kg **265°C/5Kg.

Examples of Wire Construction

An electrical wire in which the insulation consists of two polymeric layers bonded together according to the present invention was made as follows:

The inner layer of insulation (I) (i.e. nearer to the wire conductor) was a polyolefin-based material, consisting predominantly of (a) an EEA copolymer containing 15wt% EA and (b) HDPE in a weight ratio of approximately 8:2 copolymer:HDPE, with usual other additives present in smaller proportions including crosslinking promoters, stabilisers, antioxidants, pigments and process aids at a total level of 24wt%. This layer was pressure extruded onto the metallic conductor.

The outer layer of insulation (II) consisted predominantly of an ETFE copolymer, which in this example contains a crosslinking promoter at 8wt%. This outer layer was pressure extruded in a separate operation onto the pre-formed inner layer. This coated wire product was then passed through an electron beam, and received a radiation dose of 20Mrads.

In a second example a wire was made as above, in which the additives of the inner layer were added at a total level of 22.9wt% and the outer layer was comprised solely of the ETFE copolymer. This coated wire product was then passed through an electron beam, and received a radiation dose of 20 Mrads.

Demonstration of improved performance of wire as in the example above, relative to a wire of similar construction in which the inner and outer insulation layers are not bonded.

A wire of the above construction and manufacturing process (designated wire A) was compared with another crosslinked but non-bonded polyolefin/ETFE dual-walled wire (designated wire B) of the same dimensions, over a range of tests for wire robustness relevant to harsh handling and end use environments. The following results were obtained.

Example of resistance to insulation separation improvement

Method: wire size 0.75mm², wire insulation slit to depth of conductor along wire axis, slit length 50mm, 4 close-pitched wraps around a 3.60mm mandrel then unwrapped until straight, temperature 23°C

Wire Type	Result of insulation separation test
A	No spontaneous separation/delamination of core and PJ observed
B	PJ spontaneously wrinkled and separated from core

Example of scrape abrasion resistance improvement

Method: Equipment=conventional type wire scrape abrader, wire size 0.75mm²(conductor cross sectional area), blade type angled 90°, with 0.225mm radiused edge contact point, applied load 0.5Kg, stroke length 10mm, at 55 cycles/minute, temperature 23°C

Wire Type	No. of scrape cycles to abrade through PJ
A	540
B	129

Claims

1. An electrical wire or cable having insulation comprising

(I) at least a first layer of a polyolefin-based material comprising at least 20%, by weight (of the whole material composition) of a carbonyl-containing polymer (homopolymer or copolymer or terpolymer), of which polymer the or at least one constituent monomer is a carboxylic acid ester, preferably an acrylate or acetate, especially an alkyl acrylate (e.g. methyl acrylate, ethyl acrylate, propyl acrylate or butyl acrylate), the said monomer itself constituting at least 5% by weight of the said co-, or ter- polymer when used and the remainder of the said co-, or ter- polymer preferably being derived from olefinic monomer, preferably ethylene;

in contact with

(II) at least a second layer of a material containing at least 10%, by weight based on the whole material composition, of non-PVDF-based fluoropolymer or a mixture of such fluoropolymers;

wherein the said layers (I) and (II) whilst in contact with each other have been subjected to cross-linking reaction.

2. An electrical wire or cable having insulation comprising

(I) at least a first layer of a polyolefin-based formulation, of which at least 20%, preferably at least 40%, more preferably at least 60% or very preferably at least 80% of the weight of the polymeric portion of the said formulation consists of a carbonyl-containing polymer (homopolymer or copolymer or terpolymer), of which polymer the or at least one constituent monomer is a carboxylic acid ester, preferably an acrylate or acetate, especially an alkyl acrylate (preferably methyl acrylate, ethyl acrylate, propyl acrylate or butyl acrylate), the said monomer itself constituting at least 5%, preferably at least 9%, more preferably at least 15% by weight of the said co-, or ter- polymer when used, and the remainder or the majority of the remainder of the said co-, or ter- polymer preferably being derived from olefinic monomer, preferably ethylene;

in contact with

(II) at least a second layer of another material formulation, containing at least 10%, more preferably at least 50%, very preferably at least 90%, especially 100%, by weight of the second layer, of a non-PVDF-based fluoropolymer or a mixture of such fluoropolymers; wherein the said layers (I) and (II) whilst in contact with each other have been subjected to cross-linking reaction.

3. A wire or cable according to claim 1 or 2, wherein the non-PVDF-based fluoropolymer is partially fluorinated, preferably based on ETFE or ECTFE or co- or ter-polymers of ETFE or ECTFE.

4. A wire or cable according to claim 1 or 2, wherein the non-PVDF-based fluoropolymer is a ter-polymer, preferably based on tetrafluoroethylene and/or hexafluoropropylene comonomers, preferably copolymerised with ethylene or vinylidene fluoride provided the vinylidene fluoride comonomer does not constitute greater than 50wt% of the said terpolymer.

5. A wire or cable according to claim 1 or 2, wherein the non-PVDF-based fluoropolymer is a co-polymer, preferably based on tetrafluoroethylene or hexafluoropropylene comonomers, preferably copolymerised with ethylene or vinylidene fluoride provided the vinylidene fluoride comonomer does not constitute greater than 50wt% of the said terpolymer.

6. A wire or cable according to any preceding claim, wherein the said layers (I) and (II) whilst in contact with each other have been subjected to the said cross-linking reaction sufficiently to increase the peel bond strength between the said layers to at least 3N according to ASTM 1876-95 (preferably as hereinbefore described).

7. A wire or cable according to any preceding claim, wherein the said layers (I) and (II) whilst in contact with each other have been subjected to the said cross-linking reaction sufficiently to prevent delamination of the two layers on slitting of the wire along its axis, followed by wrapping and then unwrapping the wire round a mandrel of diameter equal to twice that of the insulated wire.

8. A wire or cable according to any preceding claim, wherein the cross-linking reaction has increased the bond strength by at least 50%, preferably by at least 100%, especially by at least 500% or 1000%, compared to that between the uncrosslinked layers.
9. A wire or cable according to any preceding claim, wherein the respective layers have been brought into contact with each other prior to cross-linking of either layer and at a temperature above the melting or softening point of the polymeric material in at least one of the layers.
10. A wire or cable according to any preceding claim, wherein the polyolefin-based layer comprises a mixture of polyethylene and the said carbonyl-containing polymer.
11. A wire or cable according to any preceding claim, comprising an inner layer of the said polyolefin-based material and an outer layer of the said non-PVDF-based fluoropolymer-based material.
12. A wire or cable according to claim 11, wherein the said outer layer has been pressure-extruded onto the said inner layer.
13. A wire or cable according to any preceding claim, wherein the cross-linking reaction has been effected by radiation, preferably ionising radiation.
14. A wire or cable according to any preceding claim, comprising multiple alternating layers of the materials constituting the said layers (I) and (II).
15. A wire or cable according to any preceding claim, which contains at least one crosslinking promoter in the material of either or both of the said layers (I) and (II).
16. A wire or cable according to any preceding claim, which contains at least one crosslinking promoter in the material of either or both of the said layers (I) and (II), wherein the crosslinking promoter is a multifunctional acrylate or methacrylate ester, preferably trimethylolpropanetrimethacrylate (TMPTM), or the crosslinking promoter is a multi-functional allyl compound, preferably TAC or more preferably TAIC.

17. A wire or cable according to claim 15 or 16, wherein the cross-linking promoter has been added only to the material of the said layer (I).
18. A wire or cable according to any of the preceding claims, wherein the layer (II) is substantially transparent, preferably containing substantially only ETFE or ECTFE.
19. A method of making a wire or cable according to any of the preceding claims, comprising the steps of providing on an electrical conductor the said layers (I) and (II) in contact with each other, and subjecting the said layers while in contact with each other to the said cross-linking reaction.
20. A method according to claim 19, wherein the respective layers are brought into contact with each other (a) prior to cross-linking of either layer and (b) at a temperature above the melting or softening point of the polymeric material in at least one of the layers.
21. A method according to claim 19 or 20, wherein layer (I) is pressure extruded onto the conductor and/or layer (II) is pressure extruded over layer (I).
22. A method according to claim 19, 20, or 21, wherein layers (I) and (II) are co-extruded or tandem extruded onto the wire in a single pass of the conductor from an extrusion process pay-out device to an extrusion process take-up device.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 01/01143

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H01B7/02 H01B3/44

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 H01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	WO 00 17889 A (RAYCHEM LTD ; RODWAY GILES HENRY (GB)) 30 March 2000 (2000-03-30) page 8, line 1 - line 8; claims 1-20	1-22
X	EP 0 528 611 A (CHAMPLAIN CABLE CORP) 24 February 1993 (1993-02-24) claims 5-9; example 1	1-5, 9, 11-13, 15-22
A	US 3 650 827 A (BROWN CHESTER A ET AL) 21 March 1972 (1972-03-21) abstract; claims 1-7; figure	1-22
A	EP 0 089 226 A (CHAMPLAIN CABLE CORP) 21 September 1983 (1983-09-21) page 5, line 15 - line 22; example 1	1-22
		-/-

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
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- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

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- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
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Date of the actual completion of the international search

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORTInternat'l Application No
PCT/GB 01/01143**C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT**

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 89 00759 A (RAYCHEM LTD) 26 January 1989 (1989-01-26) claims 1-8; example 2	1-22

INTERNATIONAL SEARCH REPORT

Information on patent family members

Internat	Application No
PCT/GB	01/01143

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
WO 0017889	A 30-03-2000	AU 6101999 A		10-04-2000
EP 0528611	A 24-02-1993	US 5206459 A JP 5205536 A US 5262591 A US 5313017 A US 5262592 A		27-04-1993 13-08-1993 16-11-1993 17-05-1994 16-11-1993
US 3650827	A 21-03-1972	NONE		
EP 0089226	A 21-09-1983	US 4440973 A CA 1198488 A DE 3373160 D IL 68039 A JP 58169811 A		03-04-1984 24-12-1985 24-09-1987 31-03-1987 06-10-1983
WO 8900759	A 26-01-1989	AT 103097 T AU 606731 B AU 2087788 A BR 8807603 A CA 1323674 A CA 1319403 A CA 1307837 A DE 3888537 D DE 3888537 T EP 0371048 A EP 0368886 A EP 0376966 A FI 900112 A WO 8900758 A WO 8900760 A IL 87053 A JP 2504087 T JP 2504091 T JP 2504332 T		15-04-1994 14-02-1991 13-02-1989 10-04-1990 26-10-1993 22-06-1993 22-09-1992 21-04-1994 20-10-1994 06-06-1990 23-05-1990 11-07-1990 09-01-1990 26-01-1989 26-01-1989 15-07-1992 22-11-1990 22-11-1990 06-12-1990